How does the use of different soil mineralogical atlases impact soluble iron deposition estimates?

Elisa Bergas-Massó[1,2], María Gonçalves-Ageitos[1,2], Stelios Myriokefalitakis[3], Ron L. Miller[4] and Carlos Pérez García-Pando[1,5]

[1] Barcelona Supercomputing Center (BSC), Barcelona, Spain

[2] Universitat Politècnica de Catalunya (UPC), Barcelona, Spain

[3] Institute for Environmental Research and Sustainable Development (IERSD), National Observatory of Athens, Penteli, Greece

[4] NASA Goddard Institute for Space Studies, New York, NY, United States

[5] Catalan Institution for Research and Advanced Studies (ICREA), Barcelona, Spain

Abstract

Atmospheric deposition is the primary input of soluble iron (Fe) in remote regions of the open ocean. Fe constitutes a fundamental micro-nutrient for marine biota, affecting biogeochemical cycles and ultimately carbon uptake. Fe is emitted to the atmosphere primarily from dust sources. Fe solubility at emission is origin-dependent, being mostly insoluble when associated with dust minerals. Earth System Models (ESMs) commonly assume that dust aerosols have a globally uniform composition, neglecting known regional variations in soil mineralogy. This work assesses the implications of soil composition uncertainties on the bio-available Fe delivery to the ocean by using a state-of-the-art ESM with a detailed atmospheric Fe cycle (EC-Earth3-Iron). We run two 1-year simulations considering two different Soil Mineralogy Atlases (SMAs). Our results reveal a non-negligible impact of soil mineralogy uncertainties on soluble Fe estimates. By changing the source of information for the soil composition and updating the Fe content in minerals, we find differences of 31.5% in mineral Fe emissions and 26.7% in soluble Fe deposition budgets.

Introduction

Soluble Fe acts as an essential micronutrient for marine biota, limiting phytoplankton growth in some regions of the open ocean (i.e., High Nutrient Low-Chlorophyll regions). Ocean productivity and hence CO_2 uptake by the ocean relies on the availability of limiting nutrients. Therefore, quantifying soluble Fe deposition over the ocean is key to estimate ocean primary productivity and ultimately carbon uptake.

Atmospheric mineral dust is the dominant source of Fe deposited over the open ocean. The interaction of airborne mineral dust with the Earth system depends on the mineralogical dust composition itself, particle size distribution (PSD), or its shape, among other characteristics. Hence, the Fe in dust and its ability to dissolve

is influenced by dust mineralogy. However, Earth System Models (ESMs) usually consider dust as globally uniform, disregarding the known diversity in the sources' mineralogical composition. In particular, Fe abundance in soils is generally set to a constant ~3.5% [1], and its solubility is considered to be around 0.1% [2].

Although Fe solubility at emission is low, observations suggest that solubility is boosted downwind of the sources [3] as a result of different atmospheric chemistry processes. A primary Fe solubilization mechanism is the acidic (proton-promoted) dissolution, as at low pH values, the Fe-O bonds of iron oxides weaken, favoring its dissolution [4]. Other pathways to soluble Fe production are organic ligand (e.g., oxalate) processing [5] and photoreductive processes.

Constraining the dust mineralogical composition in models presents challenges. Soil mineralogy maps are generated by extrapolating a limited number of mineralogical analyses of soil samples, which are particularly rare in arid and semi-arid regions. Furthermore, those mineralogical analyses use the wet sieving technique, which tampers the soil size distribution by breaking coarse particles, making it hard to estimate mineral-dependent PSD at emission. Moreover, the Fe content in some clay minerals can be highly variable depending on their origin: primary mineral characteristics and chemical weathering processes [6].

This work assesses the impacts of soil mineralogy uncertainties on soluble Fe deposition over the open ocean using a state-of-the-art ESM, EC-Earth3-Iron [7]. This model includes a detailed atmospheric Fe cycle and the ability to use two different soil mineralogy data sets [8] [9].

Methodology

EC-Earth3-Iron is used to model soluble Fe deposition. The ESM is composed of several modules representing different components of the Earth system [10]. The configuration used in this work includes the Integrated Forecast System (IFS) model, which represents atmospheric dynamics, coupled with the Tracer Model 5 (TM5), which allows for interactive simulation of atmospheric chemistry and transport of aerosols and reactive gas species [10]. EC-Earth3-Iron further considers:

- The primary emissions of both insoluble and soluble Fe forms, associated with mineral dust [11] and combustion aerosols [12] which are distributed in two different sizes: accumulation mode (<=1 μm) and coarse mode (1–10 μm) at emission.
- The atmospheric processing mechanism of Fe accounting for: proton-promoted dissolution, oxalate-promoted Fe dissolution (with oxalate calculated on-line) and photo-reductive dissolution. The emitted Fe is allocated in three different pools that account for different dissolution rates, namely fast, intermediate and slow Fe pools [13]
- The implementation of two different soil mineralogy datasets (Claquin [8] and Journet [9]).
- The use of Brittle fragmentation theory [14] to define the PSD of minerals at emission.

The two mineralogical datasets used, differ in various aspects (e.g, number of minerals considered in the different size ranges, spatial coverage, and observations used) The Claquin dataset [8], with the updates of Nickovic [15], provides mineralogical information for arid dust-source regions based on 239 descriptions of soils, for eight different minerals: illite, kaolinite, smectite, feldspars, hematite, gypsum, quartz, and calcite. The Journet dataset [9] is based on more than 700 soil descriptions. It contains information on the relative abundance of 12 minerals: quartz, feldspars, illite, smectite, kaolinite, chlorite, vermiculite, mica, calcite, gypsum, hematite and goethite. In contrast with Claquin, this dataset has global coverage. Journet [9] also provides updated values for the Fe mineral speciation, which are implemented in the EC-Earth3-Iron model along with the soil map. The Claquin dataset makes use of previously reported Fe content in [15] and [16].

To assess the differences in soluble iron deposition owed to uncertainties in the soil mineralogical information, we run two identical 1-year simulations (with a 1-year long spin-up) with the two different soil mineralogical datasets and Fe mineral speciation. We use the model configuration above-mentioned (IFS+TM5) for both simulations with observed sea surface temperature and sea ice concentration rather than interactive ocean. We nudge the simulation wind fields towards the ERA-Interim reanalysis [17], allowing short-term simulations to more closely follow the observed climate.

Results

Fe-dust emissions are higher with Claquin's mineralogy and Fe speciation in all dust source regions. Total budgets of Fe-dust emissions are 31.4 Tg/yr and 21.5 Tg/yr for Claquin and Journet simulations respectively, which is translated to a reduction of 31.5% in Fe-Dust emission with Journet's with respect to Claquin. This relative difference can be considered constant among particular regions and particle size ranges (Fig. 1).
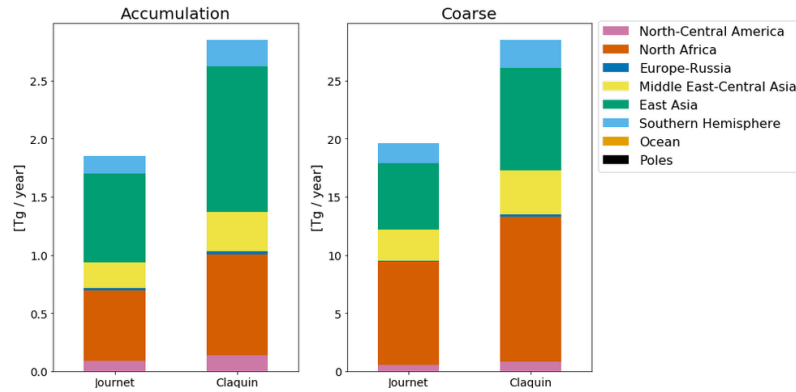

Fig. 1. Annual budget of Fe-dust emission [Tg/yr] for Journet and Claquin simulations with the accumulation mode (left) and coarse mode (right). Colors in the barplots represent the contribution to the total budget of different regions, which are based on HTAP2 regions [18].

In line with the changes in emission, soluble Fe deposition decreases for the Journet simulation compared with Claquin. This decrease is generalized over the globe but gets accentuated in the Northern Hemisphere (NH), especially in the Atlantic downwind North African dusty regions where the reduction is around 40% (Fig. 2). The sharper decrease in the relative change between Journet and Claquin over dust source regions in the NH could be explained not only by a decrease in Fe emissions, but also by higher concentrations of calcite over those regions with Journet mineralogy (more than double in almost all the NH). Higher calcite concentrations lead to a more basic atmosphere (~5% less acidic), potentially reducing the acidic processing of iron over these areas.
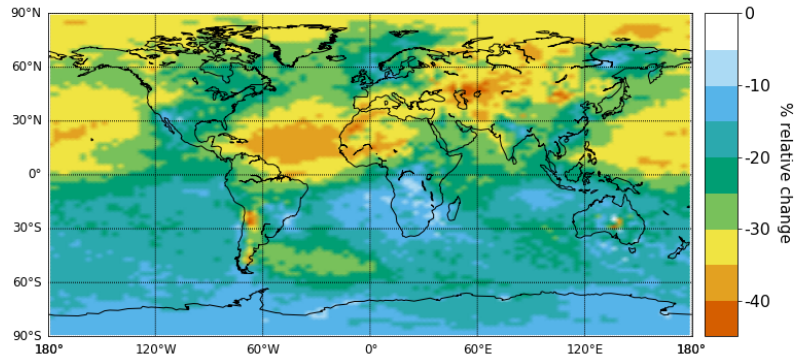

Fig. 2. Relative change [%] in soluble Fe deposition in Journet simulation with respect to Claquin simulation.

Conclusions

In this study, we assess the impact that uncertainties in mineralogical composition of soils have on soluble iron estimates. We do so by using a complex chemistry model accounting for a detailed atmospheric processing scheme, which allows us to explore non-linear effects on the resulting soluble Fe deposition budget.

Our results show that current uncertainties regarding soil composition have a significant impact on estimates of Fe-dust emissions and soluble Fe deposition. This relates to the composition of the source regions or parent soils for dust emission, from which little observations have been collected. Besides, dust composition affects aerosol pH, therefore influencing the solubilization of the iron. We detect relative differences of around 30% in Fe emissions from dust sources and about 40% in soluble Fe deposition, depending on the assumptions regarding the soil mineralogy used.

Results suggest that more emphasis should be given to constrain soil mineralogy to improve the representation of biogeochemical cycles as the Fe

cycle. To keep advancing in our understanding of how soil mineralogy influences the Fe cycle, future works will address the sensitivity of the dissolution processes towards dust mineralogy over longer periods, and with a focus on the regional impacts.

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